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MUON STUDIES OF HEAVY FERMIONS

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Full paper, invited talk to be presented at the International Conference on Magnetism 1991, Edinburg, Scotland, September 2-6, 1991

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This is an invited talk at the International Conference of Magnetism summarizing recent μSR experiments aimed at characterizing the superconducting properties of Heavy Fermion (HF) systems. Two key issues are addressed: 1) what is the symmetry of the superconducting order parameter? and 2) what is the mechanism by which electrons in HF systems pair to form the superconducting state? Both of these questions are still open at this point in time.

In a type II superconductor magnetic fields penetrate in flux vortices and the field falls off between the vortices with a characteristic length λ. Transverse field μSR experiments can measure the temperature dependence and magnitude of λ better and more easily than most other probes. UPt_3 is a hexagonal material and λ has 2 eigenvalues corresponding to an applied field perpendicular to the basal plane ($\lambda_\perp$) and parallel to the basal plane ($\lambda_\parallel$). We find that $\lambda_\perp$ is roughly linear and $\lambda_\parallel$ roughly quadratic in temperature below T_c. This corresponds to a superconducting order parameter (energy gap) which has nodes for momenta in the basal plane and along the hexagonal symmetry axis. This forms an even-parity, d-wave state

When UBe_{13} is doped with thorium to form $U_{1-x}Th_xBe_{13}$, an unusual phase diagram in the space of temperature versus Th concentration is formed. The results presented here map out this phase diagram in detail. Most interesting is a line of phase transitions which display an onset of magnetism together with a change in the superconducting state. The magnetism may be an antiferromagnetic phase with an order parameter which couples to the superconducting order parameter, or the magnetism may be inherent to the superconducting state itself. In the latter case, the superconducting order parameter breaks time-reversal symmetry.

Finally we present the results of experiments which may have identified the "smoking gun" for the mechanism behind the superconducting pairing interaction in HF systems. When UBe_{13} is doped with boron to form $U(Be_{1-x}B_x)_{13}$, the specific heat jump at the superconducting transition is greatly enhanced for

certain B concentrations, signaling an increase of the pairing strength. We discovered that this enhancement is accompanied by a change in the magnetic coupling of the conduction electrons to the f-electrons (which pair to form the superconducting state). This change in magnetic coupling is in the direction one would expect if the pairing interaction were magnetic in origin. This type of pairing mechanism had been inferred by others before, but no direct experimental evidence has yet been confirmed.

Muon Studies of Heavy Fermions

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ABSTRACT

Recent muon spin relaxation (μSR) studies have been particularly effective in revealing important properties of the unusual magnetism and superconductivity found in heavy fermion (HF) systems. In this paper μSR experiments elucidating the symmetry of superconducting order parameter in UPt_3 and UBe_{13} doped with thorium and reviewed. Also discussed is the correlation between the enhanced superconducting specific heat jump and the reduced Kondo temperature in B-doped UBe_{13}, indicating possible direct experimental evidence for a magnetic pairing mechanism in HF superconductors.

Keywords: Heavy Fermions, Superconductivity, Muon Spin Rotation

INTRODUCTION

Several important issues regarding the nature of the heavy fermion (HF) state remain after nearly a decade of study[1]. These include the extension of the isolated-impurity Kondo problem to the lattice of 4f or 5f elements (Ce, U,...), the role of small moments in the normal and superconducting properties, and the nature of the pairing interaction, including the symmetry of the superconducting order parameter. This paper will focus on the role which muon spin rotation (μSR) experiments have played in elucidating the symmetry of the HF superconducting state and the nature of the pairing interaction.

The superconducting order parameter $\Delta(\underset{\sim}{k})$ is given[2] by equations (1a) and (1b) for an even or odd parity state, respectively:

$$\Delta(\underset{\sim}{k}) = \psi(\underset{\sim}{k})\ i\sigma^{y} \qquad (S = 0) \qquad (1a)$$

$$\Delta(\underset{\sim}{k}) = i(\underset{\sim}{d}(\underset{\sim}{k}) \cdot \hat{\sigma})\sigma^{y}. \qquad (S = 1) \qquad (2a)$$

Here ψ and $\underset{\sim}{d}$ are even scaler and odd vector functions of the momentum $\underset{\sim}{k}$, and $\hat{\sigma}$ is the Pauli spin matrix. The symmetry operations of the Hamiltonian involve inversion symmetry, rotation symmetry, time-reversal invariance and gauge symmetry. An unconventional superconducting order parameter[2] breaks one of these symmetries by having, for example, odd parity, a lower rotational symmetry than the lattice or exhibiting inherent spin or orbital magnetism, which breaks time-reversal symmetry. The latter involves a $\Delta(\underset{\sim}{k})$ with both real and imaginary parts.

When $\Delta(\underset{\sim}{k})$ vanishes on lines or points of the Fermi surface, one finds power-law temperature dependences for measurements below T_c which involve thermal excitations of quasiparticles. Caution must be exercised when interpreting this as unambiguous evidence for unconventional superconductivity, however. For example, impurity scattering can lead to power-law behavior, as in nearly gapless BCS superconductivity. More unambiguous signatures of unconventional superconductivity include transitions from one superconducting phase to another, observation of magnetism associated with the superconducting state, and anisotropies in the temperature dependence of the penetration depth or critical fields, for example. Below we examine some of the evidence for unconventional superconductivity in UPt_3 and UBe_{13} doped with impurities.

II. UPt_3

Recently various studies, but principally ultrasound measurements[3-4], have demonstrated that UPt_3 is a HF superconductor which possesses multiple superconducting phases, corresponding to different representations of a multicomponent superconducting order parameter. These different phases become manifest when the degeneracy among the components of the order parameter is broken by a symmetry-breaking field, in this case either a strain field or an antiferromagnetic field[5] which breaks the hexagonal symmetry in the basal plane. The unconventionality of the superconductivity in UPt_3 has therefore been well established. The symmetry of the order parameter (parity, for example) is still controversial, however.

Recently, Broholm et.al measured[6] the temperature dependence of the penetration depth λ in UPt_3 using μSR for low applied fields ($\approx$ 180 O_e). In a hexagonal material the penetration depth λ has two eigenvalues corresponding to supercurrents in the basal plane ($\lambda_\perp$) and along the $\hat{c}$-axis ($\lambda_{||}$). In general one has $\lambda^{-2} = 4\pi e^2(n_s/m^*c^2)$, where n_s is the superfluid density and m^* the effective mass.[7] Using a standard model for $n_s(T)$, which assumes the clean-limit and weak coupling, Broholm et.al find $\lambda \sim T^\alpha$, where $\alpha = 1.3 \pm 0.1$ for $\underset{\sim}{H} \parallel \hat{c}$ and $\alpha = 2.4 \pm 0.2$ for $\underset{\sim}{H} \parallel \hat{a}$ ($\underset{\sim}{H}$ in the basal plane). The analysis yields $\lambda_{||}(0) = 6920 \pm 40$ Å and $\lambda_\perp(0) = 7200 \pm 100$ Å, corresponding to an effective mass which is roughly isotropic and about 270 times the electron mass.

The roughly linear temperature dependence for $\underset{\sim}{H} \parallel \hat{c}$ is consistent with a line of nodes in the basal plane for $\Delta(\underset{\sim}{k})$. For strong spin-orbit coupling, this implies an even-parity state.[2] The combined linear and quadratic temperature dependence is consistent with an order parameter given by $\underset{\sim}{d}(\underset{\sim}{k}) = k_z(k_x + ik_y)$, which is even parity, time-reversal violating and possesses a line of nodes in the basal plane ($k_x^2 + k_y^2 = 0$) and along the polar caps ($k_z^2 = 0$).

An apparent contradiction with this picture arises when one takes into account the analysis of the anisotropy in the upper critical field, however. At low temperatures Shivaram et.al found[8] that H_{c2} is smaller for $H \parallel \hat{c}$ than for

$\underline{H} \parallel \hat{a}$. Choi and Sauls showed[10] that for a p-wave (odd parity) superconductor one has $\underline{d} \cdot \underline{S} = 0$, so that assuming $\underline{d}$ is aligned along the axis of symmetry, one has pair breaking when $\underline{H} \parallel \underline{d} \parallel \hat{c}$, leading to a reduced H_{c2} for $H \parallel \hat{c}$. For an even-parity state one has pairbreaking for all field directions. Choi and Sauls thus conclude that UPt_3 is an odd-parity superconductor. A contradiction with the μSR data therefore occurs if the high-field (H_{c2}) and low-field (μSR) measurements can be directly compared, which may not be the case.

IV UBe_{13} doped with Th

Substitutions of Th for U in $U_{1-x}Th_xBe_{13}$ produce[11] another phase transition at T_{c2} below the superconducting transition at T_{c1} for $0.019 \leq x \leq 0.043$. Recently, a more complete phase diagram[12] for this system has been deduced (Fig. 1), wherein the transitions below T_{c2} are second-order (continuous order parameter) and are accompanied by the onset of mean-field, small-moment (10^{-2} - 10^{-3} μ_B/U-atom) magnetic correlations. The onset of this weak magnetism is illustrated in Fig. 2, where the measured zero-field μSR linewidth σ_{KT} is unchanged below T_{c1}, but rises smoothly below T_{c2}. The enhanced linewidth $\sigma_e^2(T) = \sigma_{KT}^2(T) - \sigma_{KT}^2(T_{c2})$ below T_{c2} is due to electronic magnetism, which increases in magnitude[12] as the Th concentration is increased for x = 1.93, 2.45 and 3.55 percent. (See Table I). Combined μSR and specific heat[13] measurements show that there are steep phase boundaries near x = 0.019 and x = 0.043, separating magnetic from non-magnetic regions. The fact that within errors the transitions at T_{c2} begin and terminate on the line of superconducting phase transitions at T_{c1} means that the order parameters for the two phases must be strongly coupled.

The nature of the phase below T_{c2} remains controversial. Early ultrasonic attenuation studies[14] are consistent with itinerant antiferromagnetism (AFM);

theoretical studies have also suggested a spin-density-wave (SDW) state.[15] Small local moments[1,16] associated with the Th sites or U sites have also been proposed, as well a transition to a second superconducting phase possessing orbital or spin magnetism.[17] If the transition at T_{c2} were associated with local "Kondo holes" on the Th sites, one would expect the dipolar linewidth $\sigma_e(0)$ to scale as the Th concentration x, which is not seen (see Table I). Thus, this possibility can be excluded.

The observation of both electronic magnetism and a large specific heat jump ΔC below T_{c2} (comparable to that at T_{c1}) suggest only two plausible possibilities for the second phase: either an AFM phase transition accompanied by a change in the superconducting state, or a transition to a magnetic (time-reversal-violating) superconducting phase. A third possibility, that there is only an AFM transition and no change in the superconducting state, seems to be precluded by the small associated moment and the large ΔC at T_{c2}.[12]

The possibility of a magnetic superconducting phase below T_{c2} is suggested by the correlation between the μSR linewidths and the slopes of the lower critical field H_{c1} below T_{c2} (see discussion below). For $x < 0.019$ or $x > 0.043$ $H_{c1}(t)$ shows[12] a single quadratic temperature dependence: $H_{c1}(t) \propto n_s/m^* \propto (1-t^2)$, where $t = T/T_c$. However, for $0.019 < x < 0.043$, two regions of quadratic temperature dependence are observed in $H_{c1}(t)$, one below and one above T_{c2}. These data are presented in Table I, where $H_{c1}{}^L(0)$ and $H_{c1}{}^H(0)$ are the slopes of $H_{c1}(t)$ below and above T_{c2}, respectively. Because the t^2 dependence is expected for a change in n_s and is observed both above and below T_{c2}, it seems plausible that the change in slope at T_{c2} is due to a change in n_s and not m^*. If m^* changed at T_{c2} it would have to change abruptly and not evolve significantly in temperature, which is not likely.

The fact $\sigma_e(0)$ and the slope $H_{c1}{}^L(0)$ both increase with x (Table I) might be explained by recent theoretical models[18] which describe the production of orbital currents when electrons scatter off non magnetic impurities, thereby distorting the superconducting order parameter in a complex superconducting phase. The induced currents are proportional to n_s and produce dipolar fields proportional to σ_e. A sublinear dependence of σ_e on n_s would be expected if the field sensed by the muon, averaged over the sample volume, were nearly random in direction and magnitude. This is indicated by the roughly square-root correlation seen in Table I. It remains a mystery, however, why only Th impurities induce the phase diagram and general behavior described above. Other non-magnetic impurities just suppress T_c monotonically.

IV UBe$_{13}$ doped with B

When UBe$_{13}$ is doped with B producing $U(Be_{1-x}B_x)_{13}$ T_c is changed only slightly, but ΔC at T_c can be drastically enhanced,[19] depending on the B concentration. Fig. 3 shows a comparison[20] of C/T for x = 0 and 0.0023. In addition to the much larger ΔC, the linear coefficient of specific heat γ is also larger for the x = 0.0023 sample. Beyermann et.al have shown[21] that the enhanced ΔC is largest for B concentrations around x = 0.0023. Also, the Kondo temperature (as reflected in the shoulder in C/T below 6 K in UBe$_{13}$) is reduced in the B-doped materials. High temperature susceptibility measurements[21] give an increased effective moment in the doped material compared to UBe$_{13}$, showing a tendency toward localization of the f-moment. This is consistent with a reduced value of T_K.

One possible explanation for the enhanced ΔC in B-doped UBe$_{13}$ was thought to be magnetic correlations, as in the case for Th-doping discussed above. This possibility has been eliminated by recent μSR measurements,[20] which show no enhanced linewidth below T_c for B-doped UBe$_{13}$. This, plus the narrowness of the specific heat anomaly, indicates that only a single transition with an enhanced ΔC and γ exists in the B-doped material.

The quantity ΔC is given by $\Delta C = \beta\gamma T_C$, where β measures the strength of the pairing interaction. The dramatically larger ΔC for B-doped UBe_{13} cannot be explained solely by a larger density of states γ, and so reflects an increase in the coupling strength β.[20] The intriguing possibility therefore exists that B-doping reduces T_K, leading to an increased pairing strength, thus providing possible direct experimental evidence for a magnetic pairing mechanism in UBe_{13}. For moderately strong coupling $\beta = 1.43\ [1 + 53(T_C/\omega_0)^2 \ln\ (\omega_0/3T_C)]$, where ω_0 is the characteristic boson frequency for the pairing interaction.[22] An increased value of β is consistent with a decrease in ω_0, which in turn correlates with a reduced value of T_K. Determining the relative change in ΔC between the pure and B-doped UBe_{13} is somewhat model dependent because the shape of the specific heat curves changes as well. Preliminary estimates of β using a value of γ which conserves entropy[20] below T_C give $\beta \approx 1.5$ for UBe_{13} and $\beta \geq 2.5$ for $x = 0.0023$, showing a significant enhancement. These values correspond to $\omega_0 \approx 2 - 4$ meV and < 0.7 meV, respectively. The data are therefore qualitatively consistent if the superconducting pairing interaction is driven largely by spin fluctuations. In this regard it has been shown[23] in UBe_{13} that pressure increases T_K and produces a reduced ΔC, yielding further evidence for this hypothesis.

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Table I

x(%)	$H_{c1}^{L}(0)$	$H_{c1}^{H}(0)$	$\sigma_e(x)/\sigma_e(1.93)$	$[H_{c1}^{L}(x)/H_{c1}^{L}(1.93)]^{1/2}$
0.00	----	4.32	-----------	-----------
0.66	----	3.27	-----------	-----------
1.01	----	2.64	-----------	-----------
1.93	3.79	2.28	1.00	1.00
2.45	4.91	2.89	1.11 ± 0.06	1.14 ± 0.07
3.55	5.59	3.53	1.31 ± 0.07	1.21 ± 0.07

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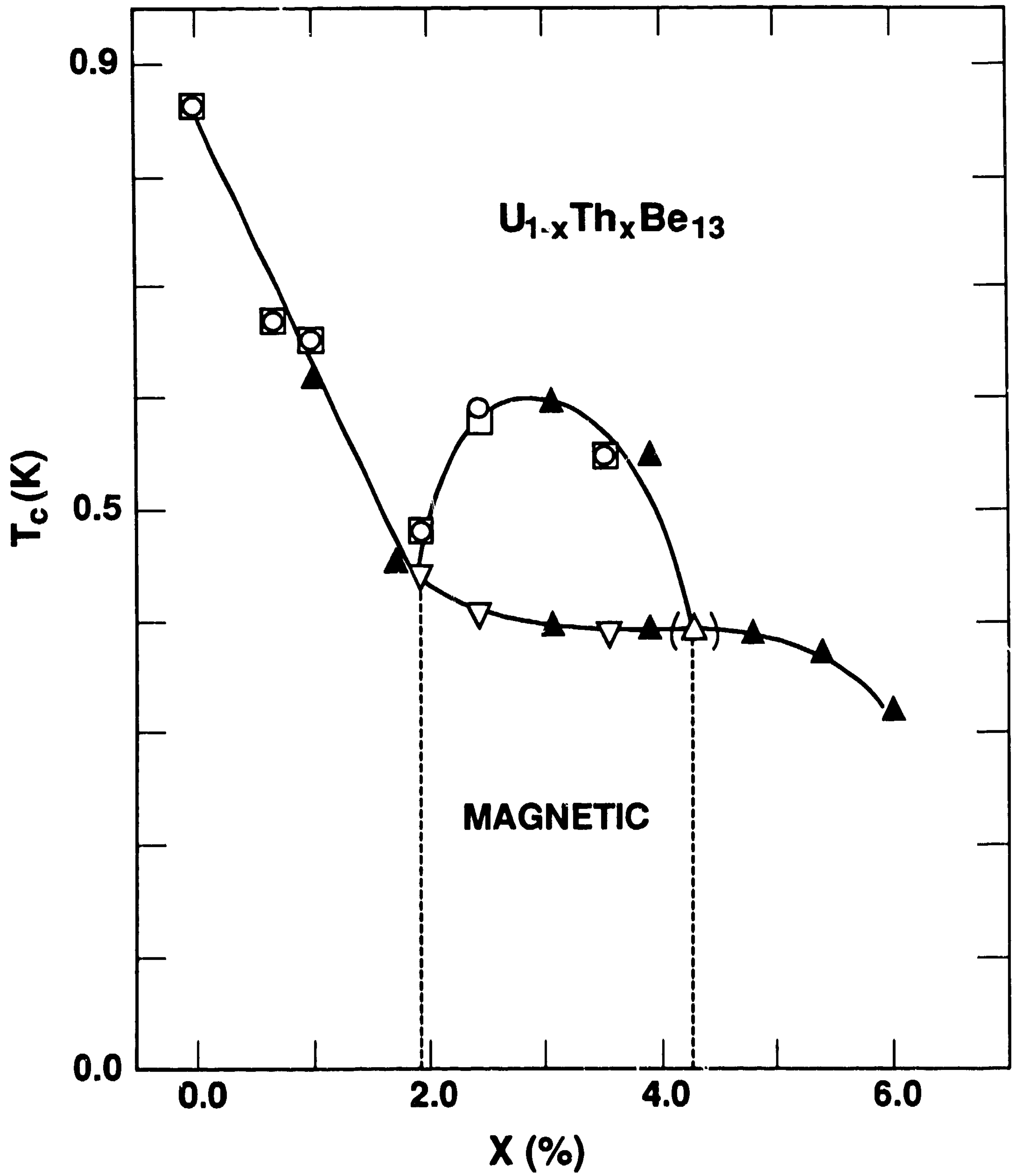


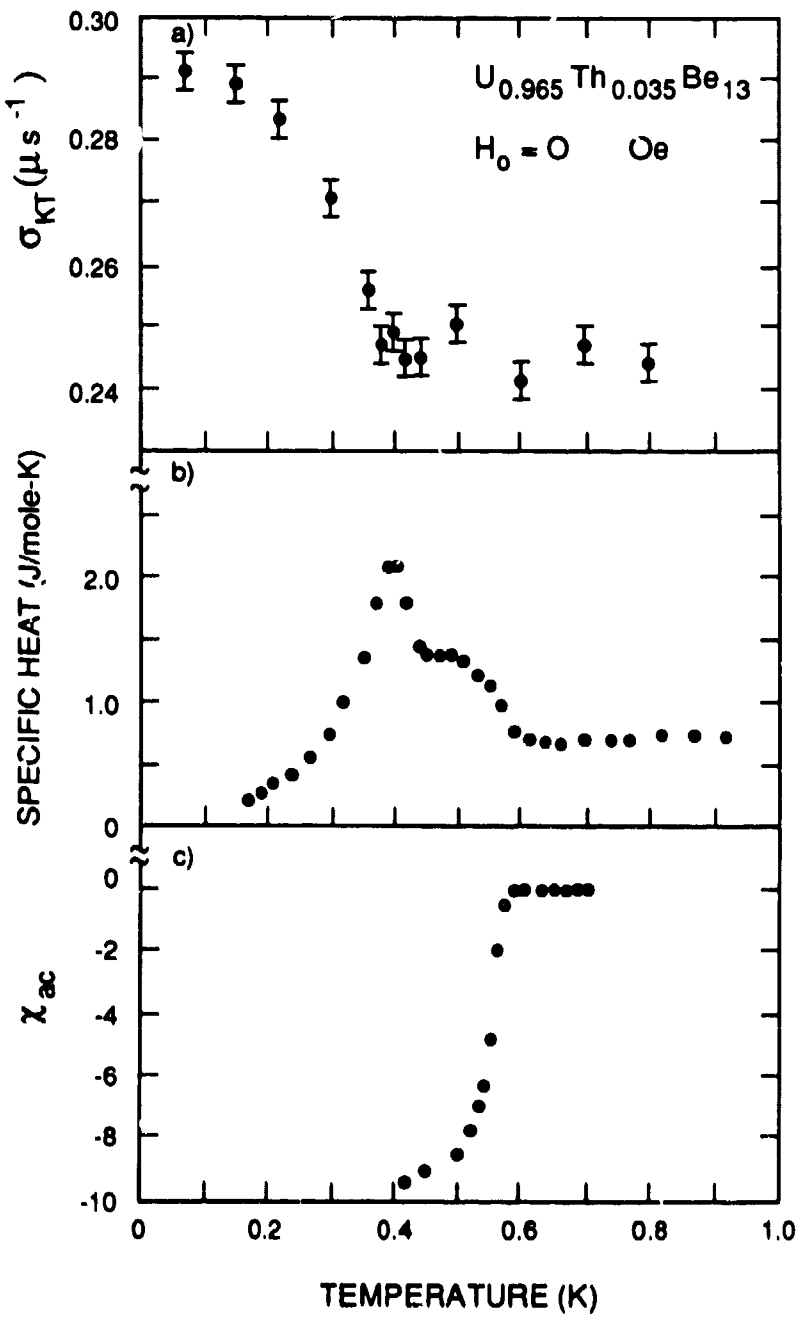
Fig 1

Figure Captions

Fig. 1 Phase diagram for $U_{1-x}Th_xBe_{13}$. Symbols, defined in Ref. 12, refer to susceptibility, specific heat and magnetization measurements.

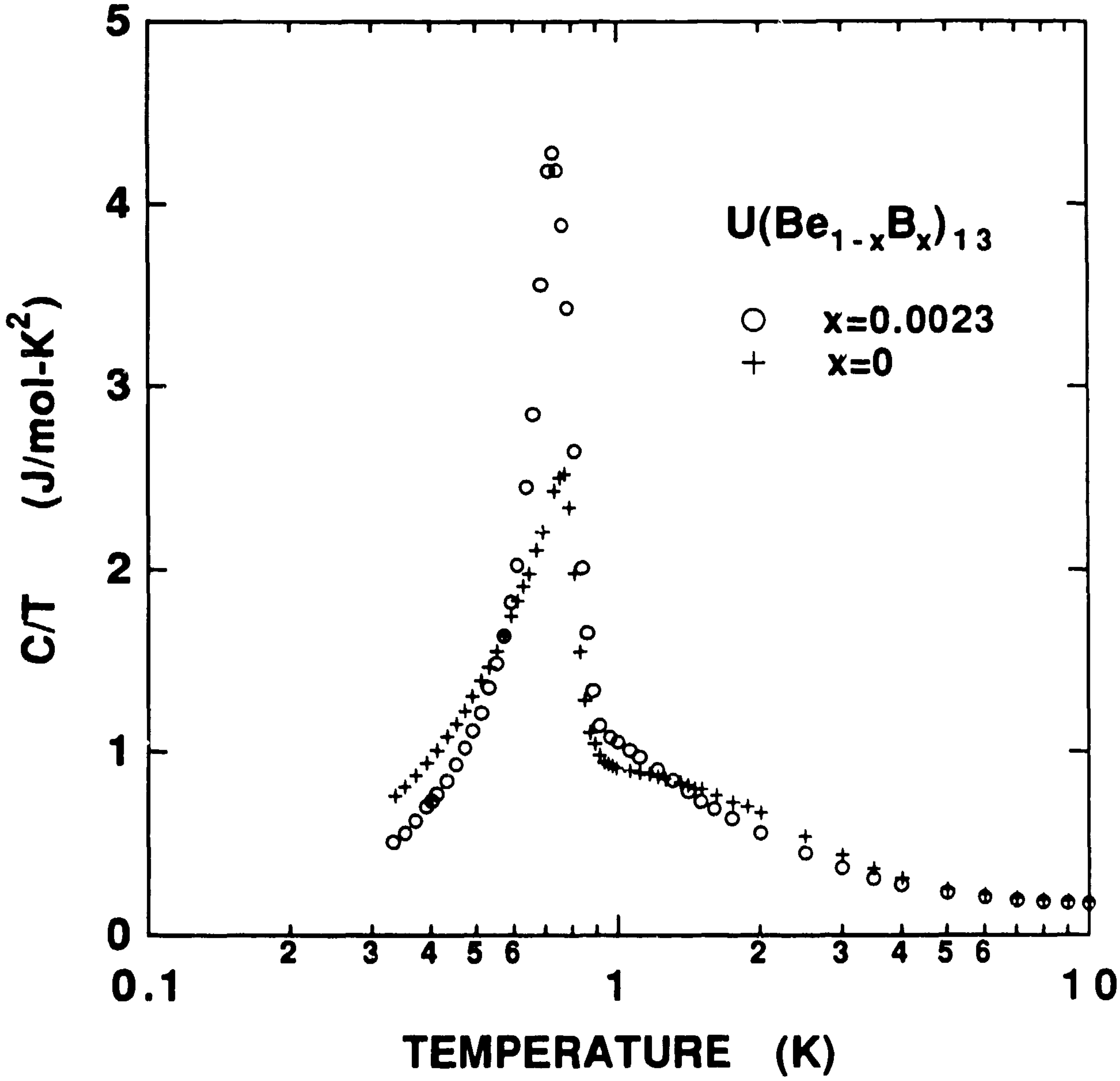
Fig. 2 Temperature dependence of (a) zero-field μSR linewidth σ_{KT}, (b) specific heat and (c) ac susceptibility in $U_{0.965}Th_{0.035}Be_{13}$.

Fig. 3 Temperature dependence of specific heat per Kelvin in $U(Be_{1-x}B_x)_{13}$.



a)
b)
c)
$U_{0.965}Th_{0.035}Be_{13}$
H_o = O Oe
$\sigma_{KT}(\mu s^{-1})$
SPECIFIC HEAT (J/mole-K)
χ_{ac}
0.30
0.28
0.26
0.24
2.0
1.0
0
0
-2
-4
-6
-8
-10
0
0.2
0.4
0.6
0.8
1.0
TEMPERATURE (K)

Fig. 2



5
4
3
2
1
0
C/T (J/mol-K^2)
$U(Be_{1-x}B_x)_{13}$
x=0.0023
x=0
0.1
2
3
4
5
6
1
2
3
4
5
6
10
TEMPERATURE (K)

Fig 3